The thickness of the metallic core depends on the metallic material used and is, for example, in the case of ~~aluminium~~ aluminum from 20 to 100 nm, preferably from 40 to 60 nm.

The thickness of the SiO_y layer (a) is generally from 20 to 500 nm, preferably from 100 to 500 nm.

The thickness of the SiO_x layer (b) is generally from 5 to 200 nm, preferably from 5 to 100 nm.

The thickness of the SiO_z layer (c) is generally from 1 to 100 nm, preferably from 5 to 100 nm, especially from 10 to 50 nm.

Preparation of the pigments according to the invention will be explained in greater detail hereinbelow with reference to ~~aluminium~~ aluminum as metal:

The ~~aluminium~~ aluminum flakes coated with SiO_x and/or SiO_y can in principle be obtained by means of a method comprising the following steps (EP-B-990 715):

a) ~~vapour~~vapor-deposition of a separating agent onto a carrier to produce a separating agent layer,
b) ~~vapour~~vapor-deposition of an Al layer onto the separating agent layer,
c) optionally, ~~vapour~~vapor-deposition of an SiO_y layer onto the Al layer,
d) ~~vapour~~vapor-deposition of an SiO_x layer onto the Al layer or, if present, onto the SiO_y layer, wherein $0.95 \leq y \leq 1.80$, especially $1.0 \leq y \leq 1.80$, very especially $1.1 \leq y \leq 1.50$,
e) optionally, ~~vapour~~vapor-deposition of an SiO_y layer onto the SiO_x layer,
f) dissolution of the separating agent layer in a solvent,
g) separation of the SiO_x-coated ~~aluminium~~ aluminum flakes from the solvent.

The SiO_y layer is preferably ~~vapour~~vapor-deposited from a ~~vaporiser~~ vaporizer in which a charge comprising a mixture of Si and SiO_2, SiO_y and mixtures thereof is present.

The SiO_x layer is produced by ~~vaporising~~ vaporizing silicon in the presence of oxygen, it being possible to control, precisely, the ratio of silicon to oxygen at a specific ~~vapour~~vapor-deposition rate by adjusting the partial pressure of the oxygen (see, for example, EP-A-803 549).

The method mentioned hereinbefore makes available SiO_x-coated or SiO_y/SiO_x-coated ~~aluminium~~ aluminum flakes that have a high degree of plane parallelism and a defined thickness in the region of ± 10 %, preferably ± 5 %, of the average thickness.

The SiO_y layers are obtained by heating a preferably stoichiometric mixture of fine silicon and quartz (SiO_2) powder in a ~~vaporiser~~ vaporizer described, for example, in DE 4 342 574 C1 and in US-A-6 202 591 to more than 1300°C under a high vacuum. The reaction product is silicon monoxide gas, which is directed *in vacuo* directly onto the passing carrier, on which it is condensed as SiO. It is also possible to use non-stoichiometric mixtures. The charge present in the ~~vaporiser~~ vaporizer comprises a mixture of Si and SiO_2, SiO_y, and mixtures thereof, the particle size of the substances reacting with one another (Si and SiO_2) advantageously being less than 0.3 mm. The weight ratio of Si to SiO_2 is advantageously in the range from 0.15:1 to 0.75:1 (parts by weight); preferably, a stoichiometric mixture is present. SiO_y present in the ~~vaporiser~~vaporizer ~~vaporises~~ vaporizes directly. Si and SiO_2 react at a temperature of more than 1300°C to form silicon monoxide ~~vapour~~vapor.

The separating agent ~~vapour~~vapor-deposited onto the carrier in step a) may be a lacquer (surface coating), a polymer, such as, for example, the (thermoplastic) polymers, in particular acryl- or styrene polymers or mixtures thereof, as described in US-B-6,398,999, an organic substance soluble in organic solvents or water and ~~vaporisable~~ vaporizable *in vacuo*, such as anthracene, anthraquinone, acetamidophenol, acetylsalicylic acid, camphoric anhydride, benzimidazole, benzene-1,2,4-tricarboxylic acid, biphenyl-2,2-dicarboxylic acid, bis(4-hydroxyphenyl)sulfone, dihydroxyanthraquinone, hydantoin, 3-hydroxybenzoic acid, 8-hydroxyquinoline-5-sulfonic acid monohydrate, 4-hydroxycoumarin, 7-hydroxycoumarin, 3-hydroxynaphthalene-2-carboxylic acid, isophthalic acid, 4,4-methylene-bis-3-hydroxynaphthalene-2-carboxylic acid, naphthalene-1,8-dicarboxylic anhydride, phthalimide and its potassium salt, phenolphthalein, phenothiazine, saccharin and its salts, tetraphenylmethane, triphenylene, triphenylmethanol or a mixture of at least two of those substances. The separating agent is preferably an inorganic salt soluble in water and ~~vaporisable~~ vaporizable *in vacuo* (see, for example, DE 198 44 357), such as sodium chloride, potassium chloride, lithium chloride, sodium fluoride, potassium fluoride, lithium fluoride, calcium fluoride, sodium ~~aluminium~~ aluminum fluoride and disodium tetraborate.

Usually, step f) is carried out at a pressure which is higher than the pressure in steps a) to e) and lower than atmospheric pressure.

The (movable) carrier preferably comprises one or more continuous metal belts, with or without a polymer coating, or one or more polyimide or polyethylene terephthalate belts. The movable carrier may furthermore comprise one or more discs, cylinders or other rotationally symmetrical bodies, which rotate about an axis.

The SiO_x-coated ~~aluminium~~ aluminum flakes are separated from the separating agent solvent preferably by washing-out and subsequent filtration, sedimentation, centrifugation, decanting or evaporation. Furthermore, the SiO_x-coated ~~aluminium~~ aluminum flakes may, after washing-out of the dissolved separating agent contained in the solvent, be frozen together with the solvent and subsequently subjected to a process of freeze-drying, whereupon the solvent is separated off as a result of sublimation below the triple point and the dry flakes remain behind in the form of individual plane-parallel structures.

The condensed silicon suboxide corresponds to the formula SiO_y wherein $0.95 < y \leq 1.8$, preferably wherein approximately $1 \leq y \leq 1.5$, y values of less than 1 being achieved by means of an excess of silicon in the ~~vaporiser~~vaporizer material. SiO_y wherein $y > 1.0$ is obtained by ~~vaporising~~vaporizing SiO in the presence of oxygen. Except under an ultra-high vacuum, in industrial vacuums of a few 10^{-2} Pa ~~vaporised~~vaporized SiO always condenses as SiO_y wherein $1 \leq y \leq 1.8$, especially wherein $1.1 \leq y \leq 1.8$, because high-vacuum apparatuses always contain, as a result of gas emission from surfaces, traces of water ~~vapour~~vapor which react with the readily reactive SiO at ~~vaporisation~~vaporization temperature.

As a result of the so-called reactive ~~vaporisation~~ vaporization of SiO in a pure oxygen atmosphere it is possible to obtain, for example, $SiO_{1.5}$ layers which do not absorb in the visible range and the refractive index of which at 550 nm is 1.55 (E. Ritter, J. Vac. Sci Technol. 3 (1966) 225).

Substantially absorption-free layers are obtained if the growing SiO_y layer is irradiated with UV light during the ~~vapour~~vapor-deposition procedure (DE-A-1 621 214).

In detail, a salt, for example NaCl, followed successively by layers of ~~aluminium~~aluminum, optionally SiO_y and SiO_x are ~~vapour~~vapor-deposited onto a carrier, which may be a continuous metal belt, passing by way of the ~~vaporisers~~ vaporizers under a vacuum of < 0.5 Pa. The thicknesses of salt ~~vapour~~vapor-deposited are about from 20 to 100 nm, preferably from 30 to 60 nm, those of ~~aluminium~~aluminum from 20 to 100 nm, those of SiO_y from 20 to 500 nm and those of SiO_x from 5 to 200 nm. On its further course, the belt-form carrier, which is closed to form a loop, runs through dynamic vacuum lock chambers of known mode of construction (cf. US 6 270 840) into a region of from 1 to 5×10^4 Pa pressure, preferably from 600 to 10^9 Pa pressure, and especially from 10^3 to 5×10^3 Pa pressure, where it is immersed in a dissolution bath. The temperature of the solvent should be so selected that its ~~vapour~~vapor pressure is in the indicated pressure range. With mechanical

assistance, the separating agent layer rapidly dissolves and the product layer breaks up into flakes, which are then present in the solvent in the form of a suspension. On its further course, the belt is dried and freed from any contaminants still adhering to it. It runs through a second group of dynamic vacuum lock chambers back into the ~~vaporisation~~ vaporization chamber, where the process of coating with separating agent and product layer is repeated.

The suspension then present in both cases, comprising product structures and solvent, and the separating agent dissolved therein, is then separated in a further operation in accordance with a known technique. For that purpose, the product structures are first concentrated in the liquid and rinsed several times with fresh solvent in order to wash out the dissolved separating agent. The product, in the form of a solid that is still wet, is then separated off by filtration, sedimentation, centrifugation, decanting or evaporation and dried.

Separating off the plane-parallel structures after washing-out at atmospheric pressure can be carried out under gentle conditions by freezing the suspension, which has been concentrated to a solids content of about 50 %, and subjecting it in known manner to freeze-drying at about -10°C and 50 Pa pressure. The dry substance remains behind as product, which can be subjected to the steps of further processing by means of coating or chemical conversion.

Instead of using a continuous belt, it is possible to produce the product by carrying out the steps of ~~vapour~~vapor-deposition of separating agent and SiO_x, Al and SiO_y, of dissolution, and of drying the carrier, in an apparatus having a rotary body, in accordance with DE-A-199 52 032. The rotary body may be one or more discs, a cylinder or any other rotationally symmetrical body.

If, under industrial vacuums of a few 10^{-2} Pa, Si is ~~vaporised~~ vaporized instead of SiO_y, silicon oxides that have a less-than-equimolar oxygen content are obtained, that is to say SiO_x wherein $0.03 \leq x \leq 0.95$, especially $0.05 \leq x \leq 0.5$, very especially $0.10 \leq x \leq 0.3$, which have astonishingly high stability to oxidation along with a high refractive index, even in thin layers. Heating in the presence of oxygen at from 150 to 500°C, preferably from 175 to 300°C, unexpectedly results in a very thin, for example approximately 20 nm thick, superficial silicon dioxide layer, which is a very convenient method of producing structures having the layer sequence $Al/SiO_x/SiO_2$. If thicker silicon dioxide layers are desired, they may conveniently be produced, as described above, by means of ~~vapour~~vapor-deposition of SiO_y and oxidative heat treatment thereof or by wet-chemical coating of the flakes with SiO_2.

For oxidative heat treatment, air or some other oxygen-containing gas is passed through the platelets, which are, for example, in the form of loose material or in a fluidised bed, at a temperature of more than 200°C, preferably more than 400°C to oxidise the SiO_y.

Accordingly, an SiO_z layer can be produced as described above, by means of ~~vapour~~vapor deposition of SiO_y and optionally oxidative heat treatment thereof.

For wet-chemical coating of the flakes with SiO_2, the following method, for example, may be used: a soda waterglass solution is metered in to a suspension of the material being coated, which suspension has been heated to about 50-100°C, especially 70-80°C. The pH is maintained at from 4 to 10, preferably from 6.5 to 8.5, by simultaneously adding 10 % hydrochloric acid. After addition of the waterglass solution, stirring is carried out for 30 minutes (see, for example, WO98/53011).

A first preferred embodiment of the present invention relates to pigments whose particles have a length of from 2 μm to 5 mm, a width of from 2 μm to 2 mm and a thickness of from 50 nm to 1.5 μm and a ratio of length to thickness of at least 2 : 1, the particles having a core of ~~aluminium~~aluminum having two substantially parallel faces, the distance between which is the shortest axis of the core, comprising

(b), on one parallel face of the core, an SiO_x layer wherein $0.03 \leq x \leq 0.95$, and

(c), on the SiO_x layer, an SiO_2 layer wherein $0.03 \leq x \leq 0.95$, especially wherein $0.05 \leq x \leq 0.5$, very especially wherein $0.10 \leq x \leq 0.30$.

The thickness of the ~~aluminium~~aluminum core in that embodiment is from 20 to 100 nm, preferably from 40 to 60 nm. The thickness of the SiO_x layer (b) in that embodiment is from 5 to 200 nm, preferably from 5 to 100 nm. The thickness of the SiO_2 layer (c) in that embodiment is from 5 to 100 nm, preferably from 10 to 50 nm.

A second preferred embodiment of the present invention relates to pigments whose particles have a length of from 2 μm to 5 mm, a width of from 2 μm to 2 mm and a thickness of from 50 nm to 1.5 μm and a ratio of length to thickness of at least 2 : 1, the particles having a core of ~~aluminium~~aluminum having two substantially parallel faces, the distance between which is the shortest axis of the core, comprising

(a), on one parallel face of the core, an SiO_y layer wherein $0.95 < y \leq 1.8$,

(b), on the SiO_y layer, an SiO_x layer wherein $0.03 \leq x \leq 0.95$, and

(c), on the SiO_x layer, an SiO_z layer, wherein $1.0 \leq z \leq 2.0$, especially $1.4 \leq z \leq 2.0$, very especially $z = 2.0$.

The SiO_z layer may, as described hereinbefore, be converted into an SiO_2 layer, as a result of which there are obtained pigments whose particles have a length of from 2 μm to 5 mm, a width of from 2 μm to 2 mm and a thickness of from 50 nm to 1.5 μm and a ratio of length to

thickness of at least 2 : 1, the particles having a core of ~~aluminium~~aluminum having two substantially parallel faces, the distance between which is the shortest axis of the core, comprising

(a), on one parallel face of the core, an SiO_y layer wherein $0.95 < y \leq 1.8$,,

(b), on the SiO_y layer, an SiO_x layer wherein $0.03 \leq x \leq 0.95$, and

(c), on the SiO_x layer, an SiO_2 layer.

The thickness of the ~~aluminium~~aluminum core in that embodiment is from 20 to 100 nm, preferably from 40 to 60 nm. The thickness of the SiO_y layer (a) in that embodiment is from 20 to 500 nm, preferably from 100 to 500 nm. The thickness of the SiO_x layer (b) in that embodiment is from 5 to 200 nm, preferably from 5 to 100 nm. The thickness of the SiO_2 layer (c) in that embodiment is from 5 to 100 nm, preferably from 10 to 50 nm.

In order to improve the physical and/or chemical properties of the pigments, further layers may be deposited.

According to R. Besold, Aluminiumpigmente für wässrige Beschichtungen – Widerspruch oder Wirklichkeit?, Farbe + Lack 97 (1991) 311 – 314, a large number of procedures, which can be divided into two groups, are known for the stabilisation of ~~aluminium~~aluminum pigments:

- adsorption of corrosion inhibitors on the pigment surface
  - phosphoric acid esters: DE-A-3020073, EP-A-170474, EP-A-133644, US-A-4,565,716, US-A-4,808,231,
  - phosphates and phosphites: US-A-4,565,716, US-A-4,808,231, EP-A-240367,
  - vanadates : EP-A-305560, EP-A-104075,
  - chromates: US-A-2,904,523, US-A-4,693,754, EP-A-259592,
  - dimeric acids: DE-A-3002175, and
- encapsulation of the pigments with a continuous inorganic protective layer:
  - SiO_2: US-A-2,885,366, US-A-3,954,496,
  - Fe_2O_3: DE-A-3003352,
  - TiO_2: DE-A-3813335,

or organic protective layer:

  - DE-A-3630356, DE-A-3147177, EP-A-477433, especially resins modified with phosphoric acid: EP-A-170474, CA-A-1,273,733, AT-A-372696, DE-A-3807588, EP-A-319971.

It is, for example, possible for the finished pigment to be subjected to subsequent coating or subsequent treatment which further increases the light, weather and chemical stability or

Please amend the paragraph that bridges pages 17 and 18 of the disclosure as follows:

Example 1

In a vacuum installation constructed in its fundamental points analogously to the installation described in US 6 270 840, there are ~~vaporised~~ vaporized from ~~vaporisers~~ vaporizers, in succession, sodium chloride (90 nm, NaCl) as separating agent at about 900°C, ~~aluminium~~aluminum (90 nm) at about from 1400 to 1500°C, SiO_y (150 nm, y = 1.0 ± 5 %) as reaction product of Si and SiO_2 at from 1350 to 1550°C and Si (120 nm, SiO_x wherein x = 0.3 ± 10 %) at a temperature of about 1600°C. ~~Vaporisation~~ Vaporization is carried out at about 0.02 Pa. For subsequent detachment of the layers by dissolution of the separating agent, the carrier on which ~~vapour~~vapor-deposition has been carried out is sprayed at about 3000 Pa with deionised water and treated using mechanical assistance by means of scrapers and using ultrasound. The NaCl passes into solution and the product layer, which is insoluble, breaks up into flakes. The suspension is removed continuously from the dissolution chamber and, at atmospheric pressure, is concentrated by filtration and rinsed several times with deionised water in order to remove Na^+ and Cl^- ions that are present. After drying and grinding, flakes are obtained whose average largest diameter is in the range from 20 to 40 µm.